Annual Report on Project Number

DE-FG07-01ID14119

LIQUID METAL BOND FOR IMPROVED HEAT TRANSFER IN LWR FUEL RODS

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Period Covered: June 4th, 2002 – June 3rd, 2003

I Introduction

The progress this past year includes: 1) A study of a simulated fission gas release from UO_2 pellets. 2) A study of the ability of pressure to push liquid metal (hereafter abbreviated as LM) into cracks in a UO_2 pellet. 3) Realistic numerical computation using temperature histories of the delay time to fission gas saturation at grain boundaries, which is a precursor to release.

<u>The effect of fission gas release into the gap</u>

The simulated fission gas release experiment was performed using porous alumina pellets (~50% open porosity) as a surrogate for high-burnup fuel. A stack of these pellets was inserted in a closed-bottom glass tube, with the pellet-glass gap filled with LM. Air, simulating fission gas, was driven out of the pellet porosity by raising the temperature. At atmospheric pressure, the LM in the gap was filled with small air bubbles. However, application of 7 atm pressure to the interior of the surrogate fuel element drove the released gas back into the pellet porosity. Save for a test-reactor irradiation, there is no way of knowing whether the plenum pressure will drive released fission gas into pores and cracks in the UO_2 pellet, or into pellet-pellet interfaces. However, the initial results obtained in this study suggest that there is a good chance that fission gas release will not drive LM from the gap.

In connection with the above study, a calculation using HEATING7 was performed to determine the magnitude and position of the peak temperature in the pellet in the event that a pocket of gas formed in the gap. The gas in a 70 μm thick hot gap was assumed to be 1 cm high and to extend 40% of the pellet's circumference. The hot spot created by this gas gap was found to be about the same temperature as if He filled the entire gap. The conclusion is that replacement of LM by fission gas in an area ~ 1.3 cm^2 will negate the benefit of LM in that position.

<u>The ability of pressurization to eliminate voids and bubbles from the gap</u>

The ability of the fuel rod's internal pressure to push LM into cracks in the pellet was investigated in the following way. Radial cracks were created in a UO_2 pellet by repeated thermal quenches. The cracked pellet was loaded in a tube filled with LM and the interior was pressurized to ~ 7 atm. After cooldown, disks cut from the pellet were observed under an optical microscope. It was found that the LM was driven into all cracks in the pellet, down to cracks as thin as 3 microns. Radial cracking always occurs in fuel pellets as they are brought up to operating temperature during startup. This experiment demonstrated conclusively that these cracks will fill with LM. The net effect will be to increase the effective thermal conductivity of the fuel, thereby lowering the fuel temperature.

Delay of the Onset of fission gas release

In the generally accepted model of fission gas released from LWR fuel, fission gas diffuses from the interior of the grains to the grain boundary, where the gas is trapped as intergranular bubbles. After the grain boundary bubbles grow large enough to touch (the condition known as "saturation"), gas release occurs through the now-open porosity. Since intragranular diffusion of fission gas depends strongly upon temperature, it is expected that with the thermal- resistance of the pellet-cladding gap eliminated by the LM filler, the incubation time to fission gas release should be considerably delayed. Using realistic fuel temperature profiles generated by FRAPCON3 (1), the approach to grain boundary saturation was calculated from a numerical solution of the quasi-stationary diffusion equation with re-solution of grain-boundary gas included. Calculated differences between time-to-saturation are as high as ~ 1 year, and as low as 1-2 days.

Each of the tasks described above is discussed in more detail in the following sections.

Other applications of the LM bond

1. The LM bond has been chosen for the NERI project (2) in which UO_2 fuel is replaced by $ZrH_{1.6}$. The thermal conductivity of the hydride fuel is much larger than that of UO_2. As a result, in helium-filled fuel elements, the largest temperature drop is across the gap. Replacement of the He bond by the LM bond eliminates this thermal resistance, with a correspondingly large decrease in the hydride fuel centerline temperature.

2. In addition, the Pb-Sn-Bi eutectic liquid alloy is being considered for the gap filler in U-Zr metal fuel. The DOE and MinAtom of Russia very recently signed an agreement to either shutdown or modify the plutonium-production reactors at Seversk and Zheleznogorsk (3). Presently, these reactors operate on a very short refueling schedule designed to produce weapons-grade plutonium in the spent fuel. A possible modification that will permit continued

operation is to double the burnup, which renders the plutonium in the spent fuel unsuitable for weapons.

However, the doubling of the burnup with the current fuel element design (stainless steel swaged onto U-Zr) risks fuel failure due to fuel swelling and excessive cladding strain. By enlarging the gap and filling it with liquid metal, this failure mode can be avoided.

3. A third interest in our laboratory's experience with liquid metals involves a study of an advanced reactor concept that uses Pb-Bi coolant with stainless-steel clad U-Zr or U-Mo fuel (4). In the event of cladding failure, the coolant directly contacts the metal fuel. The compatibility between Pb-Bi and U-Zr needs to be studied to support this fuel element concept.

II Simulation of Fission Gas Release

Porous alumina pellets were used as surrogates for the oxide pellets. A glass tube was filled with LM to ~ ¼ of the tube height under atmospheric pressure. The porous pellets were pushed into the tube one at a time. The pellet-glass radial gap was either 90 or 140 microns thick. Following loading, unfilled regions in the LM bond were observed. The top of the reservoir was capped with a pressure fitting and the assembly was pressurized to roughly 7 atm. All unfilled regions in the gap disappeared. The pressure was then released and bubbles immediately reformed. This indicates that overpressurization drives air back into the porosity and prevents any bubble formation.

In the actual UO_2 pellet case, the released fission gases cannot be driven back into UO_2 matrix. Under pressure, however, the gas may be pushed into cracks in pellets or into pellet-pellet interfaces from which it has access to the intergranular porosity in the center of the pellet.

III Liquid Metal Penetration into Cracks

To create radial cracks in the oxide, the pellet was subjected to repeated thermal cycling. Using a welding torch, the pellet was heated to red heat on one side, and then quenched in cold water. This caused extensive cracking of the pellet. After drying, the cracked pellet was pushed into a glass tube filled with LM at 140 ^{o}C to about ¼ of its height and then pressurized to 7 atm. The apparatus was held at this condition for about five minutes following which the temperature was reduced to 25 ^{o}C. The pellet was cut into several thin disks. Visual observation confirmed that liquid metal entered all visible cracks, some of which extended to the center of the pellet. Observation under an optical microscope revealed LM in cracks as narrow as 3 microns. Having LM inside the pellet increases the effective thermal conductivity, thereby decreasing fuel temperature.

IV Fission Gas Release Calculation

The objective of this work was to calculate the effect of changing from He in the fuel-cladding gap to a liquid metal on the time needed to saturate the grain boundaries with fission gas. We utilized realistic fuel temperature histories and the radial distributions calculated by the FRAPCON-3 code for a BWR 8x8 fuel assembly at linear heat ratings (LHRs) of 7, 9, 11 and 13 kW/ft as reported in a recent NRC document (5). The procedure used to generate the complete temperature distribution involved using a fuel thermal conductivity equation dependent on temperature and burnup and with four adjustable parameters. The values of the latter were chosen to reproduce the fuel temperature histories provided in the NRC report.

Three radial locations in the pellet were chosen for the calculation of the accumulation of intergranular fission gas: at the pellet center (hottest -- fission gas released fastest), at crack tip (fission gas can easily escape the pellet via the crack), and half-way between the centerline and the crack tip. The cracks are the response of the fuel to the imposed temperature profile.

Because of the very sizeable variations in reported values of fission gas diffusivities, two commonly used equations for this property were tested: D_{high} (from ref. 6) and D_{low} (from ref 7). These two diffusivities differed by roughly a factor of 20. The saturation value of gas at the grain boundary was that proposed by Dowling (8). Saturation of the grain boundaries signals the onset of gas release through the open channels formed by bubble interconnection on the grain faces. The approach to grain boundary saturation was calculated from the solution of the quasi-stationary diffusion equation (Booth sphere model) with re-solution from the grain-boundary gas included. The grain boundary gas content was followed until saturation occurred.

The most useful measure of the improvement in fission gas retention by the fuel was the delay in the time required to saturate the grain boundary for the two gap filler substances. Figure 1 shows the delay times as functions of linear heat rating for the high and low diffusivities and radial locations at the centerline and half way to the crack tip. The result for the location at the crack tip is not included because the grain-face fission gas inventory never reaches the saturation value in the time frame of interest.

The delays exhibit maxima of > 1 year at LHRs in the mid-range of the values examined. At very large or very low LHRs, the delay is as small as 1 day. The only effect of a 20-fold change in the intragranular fission gas diffusivity is to shift the maximum delay times to a different LHR; the shapes of the delay vs LHR curves are the same, as are the maximum values. The error bars represent a 20% uncertainty in the saturation value, which only results in a small uncertainty in the calculated delay times.

It is important to note that although the temperature in liquid-metal-bonded case is substantially lower than the He-filled case when the gap is open, when the gap vanishes, the temperatures in the two cases nearly equalize. Gap closure partially negates the benefit of LM in the gap. However, the difference between fuel surface and cladding inner wall temperatures with a closed gap (for a He-bonded rod) rely on conductance models that have very little experimental backing.

Full report of this calculation will be submitted to Nuclear Technology.

V Future Work

Simulation of Industrial Fuel Fabrication Methods

In a conventional fuel element fabrication, after pellets are loaded into the cladding, the rod is evacuated to remove air and subsequently pressurized with helium. To simulate the actual industrial fabrication method on a small scale, an apparatus is being constructed to allow evacuation and pressurization of a short tube with a capacity of 15 UO_2 pellets and with LM as a gap filler. The tube is heated and slightly tilted from the horizontal in order to retain the LM. In all of our previous experimentation, the cladding tube was held in a vertical position. However, this would require a very substantial modification of the fuel fabrication line in an industrial-scale operation, which loads pellets into a horizontally-mounted cladding tube. The lab-scale loading with a slightly tilted tube is designed to determine whether this near-horizontal pellet loading method is feasible. If it is, only a minor modification of industrial fabrication line will be required to fill the gap with LM. The short tube will be evacuated and pellets will be pushed through LM. Subsequently, pressure will be applied to the interior of the rod. Since there is no air in the voids, pressurization should overcome the surface tension of the voids and collapse them, resulting in a perfect LM bond for both dished and flat-end pellets.

Scale-up of the Process

A plastic cylindrical half –dome capable of accommodating a 4 m long cladding tube will be constructed. The housing will be made vacuum-tight by a gasket on the entire bottom periphery. Penetrations for vacuum and pressure operation will be installed. The housing will be tilted and the cladding tube heated by half-cylinder clam-shell elements (the Zry cladding tube need only be held at slightly over 100^{o}C to keep the LM in the molten state). To assist in gap filling, the rod will be slowly rotated as the pellets are inserted. The apparatus is shown in Fig. 2.

Quality Control

A method is needed to verify the integrity of the LM bond (i.e., the absence of void spots). Eddy-current detection was tried and did not work. A possible method is detection of radioactivity from the LM using gold activation. The method is as follows: A small quantity of gold powder is added to the amount of LM that will be used for rod fabrication. Gold dissolves in the liquid Pb-Sn-Bi eutectic alloy. The gold addition will be done in a quartz tube that will be neutron irradiated in the McClellan AFB Triga reactor near Sacramento, CA. The component metals of the LM have very low neutron absorption cross sections, but irradiated gold has a 412 keV gamma. The LM doped with irradiated gold will be used to fabricate a full-length fuel element as described in the preceding paragraph. Following cooldown, the fuel element is removed from the plastic housing and placed on a horizontal rack. A shielded Ge detector with a 1 mm by 5 mm viewing orifice is placed near the rack. The fuel element is passes by the detector, which continuously records the counts in the channel corresponding to the gold gamma ray. A drop in the count rate at a particular axial and circumferential location on the rod indicates the presence of a void. The size of the void can be measured to an accuracy corresponding to the 1 mm opening in the shielding of the detector.

VI Conclusions

Major findings are:

1. The internal rod pressure of 7 atm prevents release of gas from porous alumina pellets simulating fission gas release from UO_2 fuel. However, this aspect of fission gas behavior requires more study because fission gas may not be prevented from moving from the UO_2 matrix to the gap as easily as it is with the alumina pellets.
2. With an internal rod pressure of ~ 7 atm, LM can be pushed into cracks as narrow as 3 microns. This increases the pellet thermal conductivity.
3. Calculated differences between time-to-saturation with LM and He in the gap are as high as ~ 1 year, and as low as 1-2 days.
4. Work in the 3rd year of the project includes developing and demonstrating a method of fabricating LM-bonded fuel elements that can be accommodated by industrial-scale fuel fabricators without extensive modification of the equipment. This movement towards demonstration of industrial acceptability includes developing a method of quality control of the integrity of the LM bond.

Bibliography

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Appendix

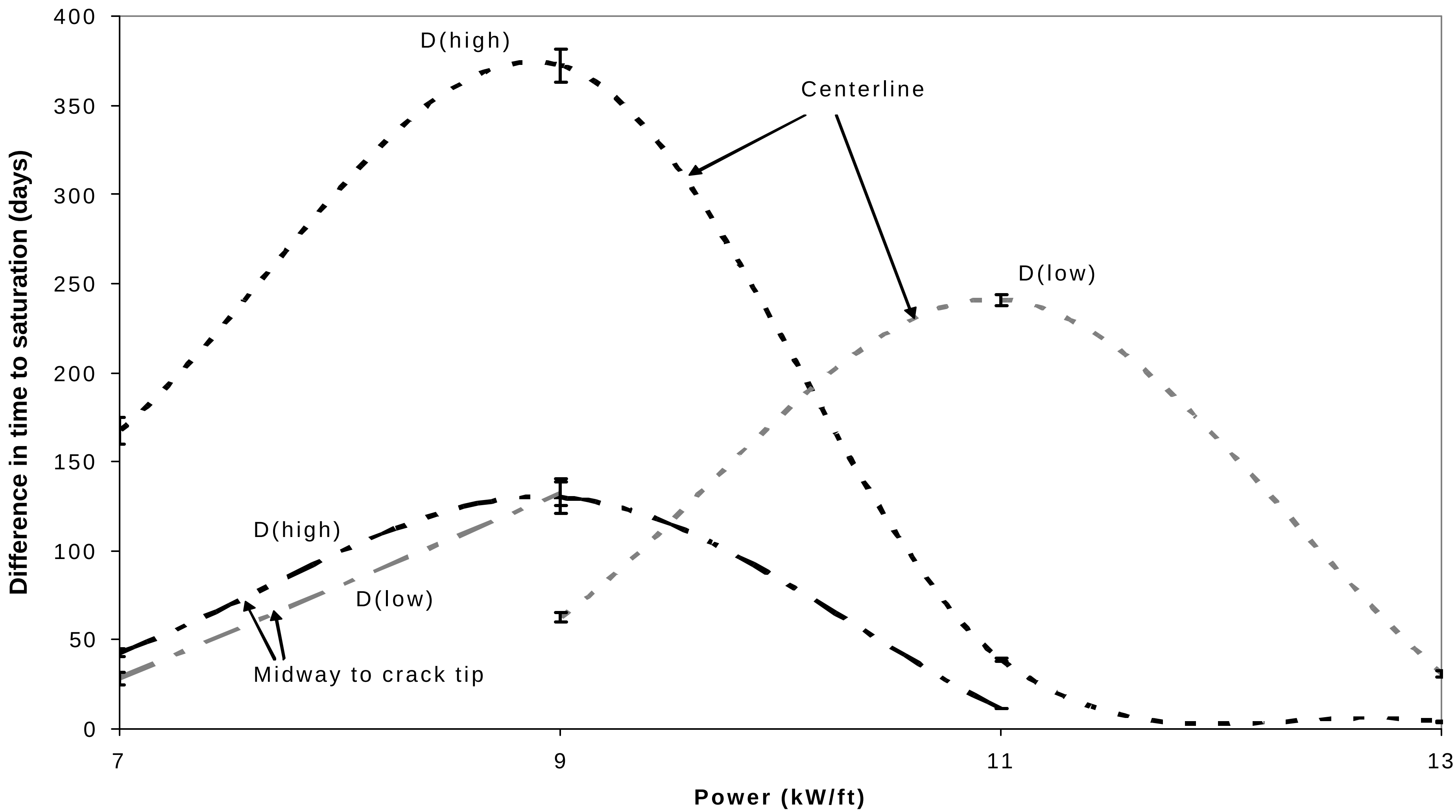

Figure 1 Difference in time to grain boundary saturation

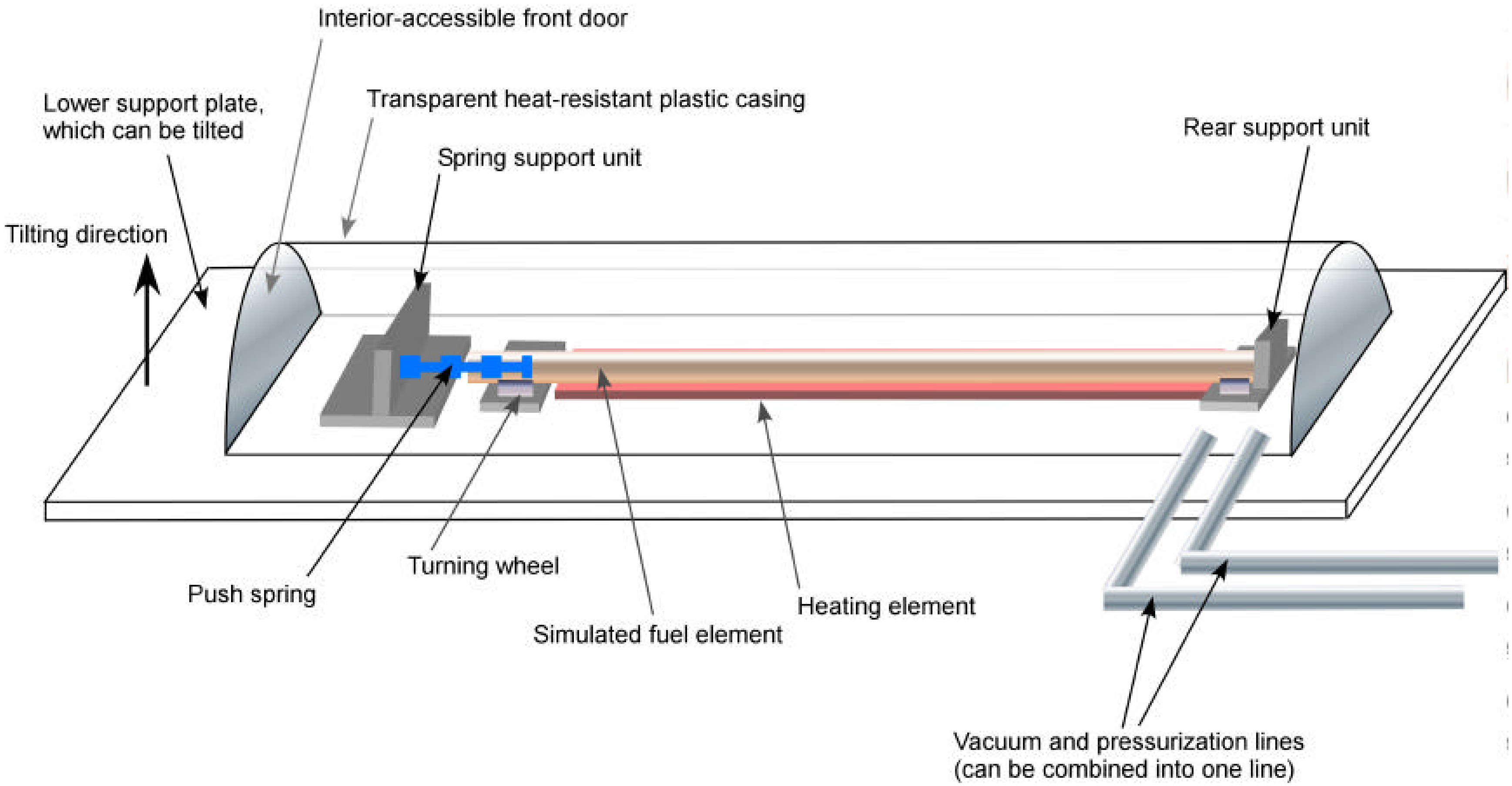

Figure 2 Full-scale fabricating apparatus